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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Elevated aerosol layer embedded with aged soot particles in a polluted urban atmosphere" *by* G. Shi et al.

Anonymous Referee #1

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General Comments

This paper presents the simultaneous collection of soot in conjunction with the measurement of several atmospheric state variables via tethered balloon. This approach is unique and yields interesting insight into how meteorology is coupled to aerosol aging dynamics. The authors should give more detailed description of the methods they used to obtain their results because some statements in the manuscript appear to be contradictory. Specifically, the authors should describe how they identify and count soot particles. They stated that 80-90% of the particles were soot at all levels of the atmosphere, which seems high. Perhaps with a more detailed description of their methods, readers can properly gauge any systematic or experimental uncertainties regarding any statistical analysis. Also, the soot particles were indirectly observed to be

in the aqueous phase even though the relative humidity (RH) never exceed %30. After discussion of these points and the points detailed below in the "Specific Comments" section, the paper should be publishable in Atmospheric Chemistry and Physics.

Specific Comments

Page 1643, Line 13: Suggest citing Molina et al.'s overview paper describing the campaign rather than an isolated result from that campaign[1]

Page 1644, Line 24: Clarify what "speculated" conditions are.

Page 1646, Line 4: Please specify what is meant by "The weather was fine".

Page 1646, Line 21-end of paragraph: I find the description aerosol sampler lacking. It is also not adequately described in Matsuki et al. How was the sampler controlled? What was the nozzle to impaction plate distance?

Page 1650, Line 19: How were the number fractions "roughly" identified? Please give detailed descriptions on how particles were identified. What kind of uncertainty and systematic error are realized in these measurements?

Page 1650, Last paragraph: That 80-95% of the particles contain soot seems high. Based on the images shown (Figure 3), I am not convinced of these numbers. What criteria did they use for soot detection? These sorts of measurements would be useful for modeling purposes, but it might be a mistake to undertake a model calculation using measurements having large uncertainties or systematic errors.

Page 1651, Line 2: Wouldn't you need to have the distance between the nozzle exit and the impaction surface to estimate the cut point? Please discuss.

Page 1651, Line 7, Page 1652, Line 7, Page 1653, Line 3: I am somewhat surprised that the particles were in the aqueous phase considering that the maximum RH was 30%. A detailed discussion of this is needed. Is it possible that the RH of the air mass was once much higher (>70%, the deliquescence point of many common aerosol

salts)? If not, then how do the authors explain the phase of the particles?

Page 1651, Line 8: Whether the particles assume a hemispherical shape or not will most likely depend on the physical properties of the particles – whether they are solid are liquid. If they are liquid, it would then depend on the contact angle of the aerosol material with the substrate. Is there any way that the authors could make a better estimation of the true geometry of the impacted particles?

Page 1651, Line 25: If the particles were too opaque to identify soot inclusions for the 1080 m layer, then how was it calculated that 90% of these particles contained soot (Page 1650, Line 23)? These are very inconsistent statements/results.

Page 1655, Line 20: Here the authors state that there are “a large number of secondary particles in the EAL besides soot particles”. What number % was this?

Page 1656, Line 8: The authors conclude that a core shell model may be a good simple model for well aged particles. Adachi et al.[2] showed that many soot particles are “embedded” and that they did not exhibit the core-shell morphology. Some discussion of these results with those obtained here is warranted.

Page 1657, Line 29: Since soot itself does not deliquesce (it is insoluble in water), suggest "deliquescence of soot containing particles".

Technical Corrections

P1642, Line 23: after “and” insert “represent an important contribution to overall PM in the urban atmosphere.” To make the sentence sound better.

Page 1642, Line 25: delete “as a carrier of black carbon” – this sounds awkward.

Page 1643, Line 1: Instead of a comma, use a dash

Page 1643, Line 5: Instead of a comma, use a dash

Page 1643, Line 7: Delete “remarkably” –sounds awkward.

Page 1644, Line 27: Did the authors mean “sensitive” rather than “sensible”?

Page 1645, Line 8: “constraining” may work better than “restraining” because it is more common in scientific literature.

Page 1646, Line 21-22: It sounds as if the impactor was used to coat the microscopy grids. I don’t think this was the case. Typically thin films are not applied using the sampler.

Page 1647, Line 16: replace “atom” with “atomic”

Page 1648, Line 2: “Barometric meter” is not a common term. Please precisely define what was used to obtain height.

Page 1650, Line 14: replace “submicron meters” with “sub micrometer”.

Page 1650, Line 17: replace “flying ash” with “fly ash”.

Page 1651, Line 11: Use of the term “more or less” is confusing here. Which is it? I think the authors meant that projected diameter of the particles is larger after impaction.

Page 1651, Line 13-14: By “geometric size” I think the authors mean “original size of the ambient particle”.

Page 1655, Line 24-25: Replace “size segregated distributions” with “size distributions”.

Page 1656, Line 2: Replace “nocturnal” with “nocturnally”.

Page 1657, Line 25, 27: Replace soot “parts” with “inclusions” or “cores”.

References: (1) Molina, L. T. et al. Air quality in North America’s most populous city - overview of the MCMA-2003 campaign. Atmospheric Chemistry and Physics 2007, 7, 2447. (2) Adachi, K.; Chung, S. H.; Buseck, P. R. Shapes of soot aerosol particles and implications for their effects on climate. Journal of Geophysical Research-Atmospheres 2010, 115.

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